CO_2 Sequestration and Recycle by Photosynthesis with Visible Light

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Abstract

Visible light-photocatalysis could provide a cost-effective route to recycle CO_2 to useful chemicals or fuels. The objective of this study is to investigate the reactivity of adsorbates, their role in the photosynthesis reaction, and their relation to the nature of surface sites during photosynthesis of methanol and hydrocarbons from CO_2/H_2O over four types of TiO_2 and CdS catalyst supported with either Al_2O_3 or MCM-41: (i) ion-exchanged metal cations, (ii) highly dispersed cations, (iii) monolayer sites, and (iv) modified monolayer catalysts. TiO_2 was selected because it is known to be highly active; CdS was selected for its photocatalytic activity in the visible light region. Al_2O_3 provides excellent hydrothermal stability. MCM-41 offers high surface area (more than 800 m^2/g), providing a platform for preparing and depositing a large number of active sites per gram catalyst. The unique structure of these ion exchange cations, highly dispersed cations, and monolayer sites provides an opportunity to tailor their chemical/coordination environments for enhancing visible-light photocatalytic activity and deactivation resistance. The year one research tasks include (i) setting up experimental system, (ii) preparing ion-exchanged metal cations, highly dispersed cations, monolayer sites of TiO_2 and CdS, and (iii) determination of the dependence of methanol activity/selectivity on the catalyst preparation techniques and their relation to adsorbate reactivity. During the first year, we purchased a Gas Chromatograph and constructed 3 reactor systems: a quartz slurry reactor, combinatory reactor, and an *in situ* infrared reactor. We configured and calibrated the light source apparatus and prepared catalysts. The quartz slurry and combinatory reactors were designed for catalyst screening. Cu/TiO_2 exhibited high activity for methanol synthesis. The *in situ* infrared was used for mechanistic studies of the reaction. *In situ* IR experiments have shown the presence of adsorbed carbonate and hydroxyl groups as well as adsorbed CH_x species. It is speculated that the product concentration remains too small to be detected by IR. Further work is underway to determine the reactive intermediates. Repeated runs are planned to insure the reproducibility of the data.

The overall goal of this research is to provide a greater predictive capability for the design of visible light-photosynthetic catalysts by in-depth understanding of the reaction kinetics and mechanism as well as by better control of the coordination/chemical environment of active sites.